

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE



In re application of :  
Makoto AKIZUKI et al. :  
Serial No. [NEW] : **Attn: APPLICATION BRANCH**  
Filed December 26, 2001 : Attorney Docket No. 2001-1897

METHOD FOR FORMING GAS CLUSTER  
AND METHOD FOR FORMING THIN FILM

**(Rule 1.53(b) Continuation of Serial  
No. 09/799,681, Filed March 7, 2001)**

**INFORMATION DISCLOSURE STATEMENT**

Assistant Commissioner for Patents,  
Washington, DC 20231

Sir:

Pursuant to the provisions of 37 CFR 1.56, 1.97 and 1.98, Applicants request consideration of ☒ the references listed on attached form PTO-1449 and/or ☐ the additional information identified below in paragraph 3. A legible copy of each reference listed on the form PTO-1449 and each U.S. patent application listed below is enclosed, except a copy is not provided for each reference previously cited by or submitted to the Patent Office in prior parent application Serial No. 09/799,681.

1a. ☒ This Information Disclosure Statement is submitted:

within three months of the filing date (or of entry into the National Stage) of the above-entitled application, **or**

before the mailing of a first Office Action on the merits or the mailing of a first Office Action after the filing of an RCE,

**and thus no certification and/or fee is required.**

1b. ☐ This Information Disclosure Statement is submitted

after the events of above paragraph 1a and prior to the mailing date of a final Office Action or a Notice of Allowance or an action which otherwise closes prosecution in the application, and thus:

(1) ☐ the certification of paragraph 2 below is provided, **or**

(2) ☐ the fee of \$180.00 specified in 37 CFR 1.17(p) is enclosed.

1c. ☐ This Information Disclosure Statement is submitted:

after the mailing date of a final Office Action or Notice of Allowance or action which otherwise closes prosecution in the application, and prior to payment of the issue fee, and thus:

**the certification of paragraph 2 below is provided, and**

**the fee of \$180.00 specified in 37 CFR 1.17(p) is enclosed.**

1d. ☐ Each item of information contained in this Information Disclosure Statement was cited in a communication from a foreign patent office in a counterpart application and this information was not received by any individual designated in 37 CFR 1.56(c) more than 30 days prior to the filing of this Information Disclosure Statement. Accordingly, this application is entitled to the protection of 37 CFR 1.704(d) with regard to the filing of this Information Disclosure Statement.

2. It is hereby certified

a. ☐ that each item of information contained in this Information Disclosure Statement was first cited in any communication from a foreign patent office in a counterpart foreign application not more than three months prior to the filing of the Statement, or

b. ☐ that no item of information contained in the Information Disclosure Statement was cited in a communication from a foreign patent office in a counterpart foreign application and, to the knowledge of the person signing the certification after making reasonable inquiry, was known to any individual designated in §1.56(c) more than three months prior to the filing of the Statement.

3. ☐ Consideration of the following additional information (including any copending or abandoned U.S. application, prior uses and/or sales, etc.) is requested.
4. For each non-English language reference listed on the attached form PTO-1449, reference is made to:
- a. ☐ a full or partial English language translation submitted herewith,
  - b. ☐ a foreign patent office search report (in the English language) submitted herewith,
  - c. ☐ the concise explanation contained in the specification of the present application at page \_\_\_\_\_,
  - d. ☐ the concise explanation set forth in the attached English language abstract,
  - e. ☒ the concise explanation set forth below or on a separate sheet attached to the reference:

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**Difference between the Japanese Patent Provisional Publication JP-A-6-2241<sup>+</sup>56 (1994) (cited reference), corresponding to US 5,527,731 (Yamamoto et al.):**

The Cited Reference: According to the cited reference, light radiation energy is used to ionize gases for surface treatment for the purpose of generating by separation, ions of a specific mass number and of a specific internal energy state, suitable for a desired surface treatment. The ions thus generated are used for surface treatment including dry etching and deposition.

According to working example 1 in the cited reference, the gases that are to be ionized include  $\text{Cl}_2$ ,  $\text{SF}_6$  and other halides, oxides, hydrides, nitrides, and inorganic complexes, these being used in the state of gas or liquid. According to working example 2 in the cited reference, cluster beams of mixed  $\text{He/SF}_6$  gases are ionized. According to working example 4 in the cited reference, gases of organic metal complexes are used as the gas to be ionized.

Claim 1: However, none of the working examples 1 through 9 in the cited reference reveals the feature of claim 1 of the present application wherein the substances in the liquid state are clustered, the clusters are generated by mixing substances in the liquid state with pressurizing gases at ordinary temperatures and pressures and then the mixture is jetted from a nozzle as a gas.

Figs. 2 and 8 in the cited reference show a system to form molecular and cluster beams. It is impossible for this technique to stably obtain cluster beams from substances in the liquid state. It is totally unclear if cluster beams can ever actually be formed with this technique.

To generate clusters in the liquid state at ordinary temperatures and pressures using the system shown in Fig. 2, it is at least necessary to mix liquid materials with He or other gaseous materials in cylinder 25. Such mixing is generally not employed. It is not considered that the specific disclosure in this cited reference is such as to be suggestive of the very different specific mixing of the present invention.

To form clusters, the supply pressure of gases (raw material) must be increased to 2 atm or above, or, as shown in working example 2 in the cited reference, 3 atm or above, whereas the vapor pressure of liquid materials at ordinary temperatures and pressures is 1 atm or below. That is, it is unrealistic to try to mix both in cylinder 25.

It is thus appropriate to assume that, despite the description in the cited reference the state of the gases may be either gaseous or liquid and that gases in the gaseous state are mixed according to the procedure revealed in the cited reference.

Furthermore, even if gaseous materials could be mixed with liquid materials in cylinder 25, the pressure of the gaseous materials in the cylinder will gradually drop as the materials are consumed. As a result, the ratio of mixture of liquid with gaseous materials in the mixed gases that are jetted from the nozzle is variable and it is thus not possible to stably maintain the condition required for the formation of clusters. Furthermore, it is difficult to generate clusters of liquid materials themselves.

This is because the strength of cluster beams depends on the ratio of vapor pressure of liquid materials to the supply pressure of gaseous materials.

In sum, the cited reference does not disclose or suggest the feature of claim 1 of the present invention that substances in the liquid state are mixed with pressurizing gases at ordinary temperatures and pressures and the mixture is clustered as it is jetted from a nozzle as gases.

Claims 2 through 10: Claims 2 through 10 of the present invention are dependent on claim 1, discussed above, and patentability is primarily based on patentability of claim 1.

With regard to claims 9 and 10, in particular, working Example 4 in the cited reference refers to film deposition using organic metal materials. This, however, does not suggest the use of cluster ions.

In the cited reference, furthermore, dissociated metallic ions are specifically generated and captured out of the generated organic metallic ions and are irradiated on to the surface of a substrate to deposit film. In contrast, in claims 9 and 10 of the present application, the generated clusters are irradiated on to the surface of a substrate without dissociation. This makes it possible to transfer kinetic energy of clusters to the substrate, with the result that a high-grade thin film is formed.

Claims 11 through 14: None of working examples 1 through 9 in the cited reference shows formation of thin film using cluster ions. However, when gaseous materials are used as raw materials, clusters can be generated and a thin film formed using a technique such as one shown in working example 2 of the reference.

However, the cited reference does not reveal that thin film is deposited by irradiating two or more chemically reactive species simultaneously.

Claims 11 through 14 of the present application go one step further in this respect by requiring that "two or more chemically reactive species, including at least one type of gas cluster ion, are irradiated to deposit thin film through reaction thereof."

A high-grade thin film can be formed at low temperatures by using kinetic energy of gas cluster ions in this reaction.


In working example 7 in the cited reference, oxygen gases are ionized to form oxides on a Si surface. This is an example of formation of thin film through oxidation of irradiated oxygen ions with Si, the constituent atom of substrates. It is thus basically different from and unsuggestive of the technology of deposition of thin film presently claimed.

Claims 15 through 18: Working example 2 in the cited reference requires a mixture of noble gases. However, mixing of noble gases with oxygen gases is not suggested.

5. [ ] A foreign patent office search report citing one or more of the references is enclosed.

Respectfully submitted,

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